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## EUROPEAN PATENT APPLICATION

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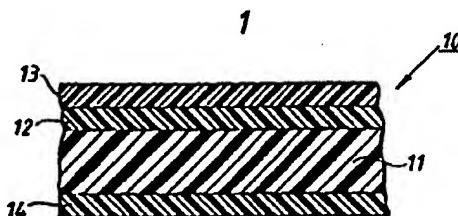
(54) Packaging material and also use of the material for manufacture of containers with good oxygen-tight properties.

(55) Packaging material for shaping into form-stable liquid-tight containers with good oxygen-tight properties through fold forming, thermo-forming or other mechanical processing for shaping of the material.

The packaging material (10) comprises a skeletal layer (11) and a barrier layer (12) attached to the skeletal layer with good adhesion. The skeletal layer (11) is composed of a mixture of plastic, preferably polyolefine, and filler to an amount of between 50 and 80% of the total weight of the mixture, and the barrier layer (12) is composed of a plastic material comprising plastic of the same type as the plastic in the skeletal layer (11), preferably polyolefine, in a mixture with plastic of another type than the one mentioned.

The plastic of the aforesaid other type in the barrier layer (12) can be an ethylene/vinyl alcohol copolymer, polyvinylidene chloride, polyamide or similar plastic with good oxygen-tight properties, preferably an ethylene/vinyl alcohol copolymer to an amount of 40% or more of the total weight of the barrier layer.

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The present invention concerns a flexible packaging material in the form of a sheet or strip for shaping into form-stable liquid-tight containers with good oxygen-tight properties through fold forming, thermo-forming or other mechanical processing for shaping of the material, with this material having a stiffening skeletal layer composed of a mixture of plastic and filler, and also a barrier layer attached to one side of the skeletal layer with good adhesion.

In addition the invention concerns the use of the packaging material for manufacture of form-stable liquid-tight containers with good oxygen-tight properties through fold forming, thermo-forming or other mechanical processing for shaping of the material.

A packaging material of the type which is described above is known through, for example, EP-A-O 353 991. A similar packaging material is also described in EP-A-O 353 496.

The known packaging material according to EP-A-O 353 991 comprises a stiffening skeletal layer composed of a mixture of either a propylene homopolymer with a melting index of under 10 according to ASTM (2.16 kg; 230 °C) and filler to an amount of between 50 and 80% of the total weight of the mixture, or of an ethylene/propylene copolymer with a melting index of between 0.5 and 5 according to ASTM (2.16 kg; 230 °C) and filler to an amount of between 50 and 80% of the total weight of the mixture, and also a layer of a material with good gas barrier properties attached to one side of the skeletal layer with good adhesion. As an individual example of such a gas barrier layer Al foil is mentioned, which is attached to the skeletal layer with the aid of a sealing layer of suitable character between them.

A problem with the known packaging material is that an Al foil, as is known, is very expensive and therefore inevitably entails high costs for production of the packaging material. An Al foil is furthermore very sensitive to tension and in practice completely inelastic and therefore breaks easily when the material is subjected to powerful tension and flexion stresses during the shaping of the material into containers. In addition according to the requirements of the authorities an Al foil which is to be used in a packaging material for containers for contents of the liquid foods type must be covered with at least one outer protective layer in order to prevent direct contact between the Al foil and the contents of the container produced, which contributes to further increasing the manufacturing costs of the material at the same time as it gives the material an unnecessarily complicated structure.

Another known packaging material of the type which is described above comprises a stiffening

skeletal layer likewise composed of a mixture of either a propylene homopolymer with a melting index of under 10 according to ASTM (2.16 kg; 230 °C) and filler to an amount of between 50 and 80% of the total weight of the mixture or of an ethylene/propylene copolymer with a melting index of between 0.5 and 5 according to ASTM (2.16 kg; 230 °C) and filler to an amount of between 50 and 80% of the total weight of the mixture, and also a homogeneous gas barrier layer of an ethylene/vinyl alcohol copolymer (EVOH) attached to one side of the skeletal layer. Since the oxygen permeability of the barrier layer is largely dependent on the barrier layer's moisture content and increases with increasing moisture contents, the known packaging material has an outer layer of plastic attached to the barrier layer, which gives the underlying barrier layer protection against the action of moisture. The skeletal layer also has an outer layer of plastic, which is preferably of the same type as the plastic in the first mentioned outer plastic layer.

This known packaging material has the advantage over the previously described known packaging material that instead of an expensive, tension-sensitive Al foil it uses a relatively cheaper ethylene/vinyl alcohol copolymer as material in the barrier layer, but it has like the previous packaging material a complicated material structure containing several different individual layers of material. The material structure is further complicated by the fact that the barrier layer of EVOH cannot be attached directly to the two surrounding layers (the skeletal layer and the moisture-protection outer plastic layer), but requires one or more intermediate binder layers in order to give the packaging material the desired good adhesion between the individual layers of material.

An aim of the present invention is therefore to give indications of a packaging material of the type described in the introduction, without resultant problems of the type inherent in the known packaging materials.

This and other aims and advantages are achieved according to the invention through the fact that the packaging material has been given the characteristic that the barrier layer is composed of a mixture of plastic of the same type as the plastic in the skeletal layer and a plastic of another type than the one mentioned.

Through the fact that the barrier layer contains the same plastic as the skeletal layer these two layers can be attached directly to each other with extraordinarily good adhesion without the use of any separate intermediate binding or sealing layer as in the known technologies. In addition the advantage is gained over the known packaging materials that the packaging material according to the invention is what is known as a homogeneous

material, i.e. a material with in all essentials the same plastic in all material layers forming part of the material, which makes the material reusable and very attractive from the production point of view. Through its easy reusability the packaging material is in addition extremely valuable from the environmental standpoint.

According to a specially advantageous embodiment of the invention the barrier layer is composed of a mixture of either 40-80 weight % EVOH and 60-20 weight % of a propylene homopolymer or an ethylene/propylene copolymer of the type described above, while the skeletal layer is composed of the same propylene homopolymer and between 50 and 80 weight % filler or of the same ethylene/propylene copolymer and between 50 and 80 weight % filler. Preferably the amount of EVOH in the barrier layer should be about 50%, which gives the barrier layer an oxygen-tightness almost in the same class as an Al foil, with the use of the minimum possible amount of EVOH, as will be explained.

Further practical and advantageous embodiments of the invention have further been given the characteristics stated in the sub-claims.

The invention will now be described and explained in greater detail with particular reference to the enclosed drawings in which

Figure 1 shows schematically a cross section of a packaging material according to a preferred embodiment of the invention,

Figure 2 explains schematically the manufacture of the packaging material in figure 1, and

Figure 3 is a graph which shows schematically how the oxygen permeability ( $O_2$  perm.) of the barrier layer varies with the proportion of EVOH in the barrier layer.

The packaging material according to the invention, which may be in the form of a sheet or strip, has been given the general reference designation 10 in figure 1. The material 10 has a stiffening skeletal layer 11 composed of a mixture of plastic and filler, and also a barrier layer 12 composed of a mixture of plastic of the same type as the plastic in the skeletal layer 11 and a plastic of another type than the one mentioned, attached to the skeletal layer 11 with good adhesion.

The barrier layer 12, which is preferably attached to the skeletal layer 11 through surface fusion of the plastic of the aforesaid same type in the respective layers, may have an outer layer 13 of plastic of the aforesaid same type. The skeletal layer 11 may have an outer layer 14 composed of plastic of the same type as in the first-mentioned outer layer 13. The two outer layers 13 and 14 are preferably also attached to the respective neighbouring layers through surface fusion of the plastic of the aforesaid same type in order to ensure good

adhesion between all layers of material forming part of the material 10.

The plastic in the skeletal layer 11 is preferably a polyolefine plastic such as polythene, polypropylene, etc., but is preferably composed of a propylene homopolymer with a melting index of under 10 according to ASTM (2.16 kg; 230 °C) or an ethylene/propylene copolymer with a melting index of between 0.5 and 5 according to ASTM (2.16 kg; 230 °C). Of these two preferred polyolefine plastics ethylene/propylene copolymer with the aforesaid melting index is the most preferred, since it retains its excellent sealing and mechanical strength properties even at low temperatures, 8 °C or lower.

The filler in the skeletal layer 11 can be any known filler in the field such as granular or flaked chalk, talc, mica, clay etc. The preferred filler is, however, chalk. The amount of filler in the skeletal layer is 50-80% of the total weight of the skeletal layer, preferably c. 65 weight %.

The barrier layer 12 is composed, as stated, of a mixture of plastic of the same type as the plastic in the skeletal layer 11 and a plastic of another type. The plastic of the aforesaid same type is thus preferably a polyolefine plastic such as polythene, polypropylene etc., but preferably a polypropylene plastic. The preferred polypropylene plastic is either a propylene homopolymer with a melting index of under 10 according to ASTM (2.16 kg; 230 °C) or an ethylene/propylene copolymer with a melting index of between 0.5 and 5 according to ASTM (2.16 kg; 230 °C). For the same reason as above, however, an ethylene/propylene copolymer with a melting index in the range mentioned is preferred. The other plastic component in the barrier layer 12 can for example be an ethylene/vinyl alcohol copolymer (EVOH), polyvinylidene chloride (PVDC), polyamide (PA) or a similar polymer with good gas barrier properties, but preferably an ethylene/vinyl alcohol copolymer (EVOH). The amount of this second plastic component in the barrier layer 12 can vary within wide limits, but is generally situated within the area 40-80% of the total weight of the barrier layer. A barrier layer composed of c. 40 weight % ethylene/vinyl alcohol copolymer (EVOH) and c. 60 weight % of an ethylene/propylene copolymer with a melting index within the region of 5-10 according to ASTM in combination with a skeletal layer 11 composed of a mixture of the same ethylene/propylene copolymer and between 50 and 80 weight % filler has in practice proved to be able to give a packaging material with oxygen-tight properties easily on a par with an Al foil, as will be explained in greater detail with particular reference to figure 3.

The two outer layers 13 and 14 are also preferably composed of a polyolefine plastic such as

polythene, polypropylene etc., preferably a polypropylene plastic which may be either a propylene homopolymer with a melting index of under 10 according to ASTM (2.26 kg; 230°C) or an ethylene/propylene copolymer with a melting index of between 0.5 and 5 according to ASTM (2.16 kg; 230°C). Preferably the two outer layers are composed of an ethylene/propylene copolymer with a melting index within the range mentioned for reasons which are explained above.

As can be seen from figure 1 the material layers forming part of the material 10 have different material thicknesses, where the thickness of the skeletal layer 11 can be between 150 and 1400 µm, the barrier layer can have a thickness of between 5 and 50 µm and the two outer layers 13 and 14 can have a thickness of between 5 and 50 µm. The thickness of the skeletal layer 11 is preferably situated in the upper part of the thickness range mentioned if the material 10 is to be shaped into a container through thermo-forming or other mechanical processing for shaping, while the thickness is preferably situated in the lower part of the range if the material is to be shaped into a container through fold forming.

Figure 3 shows a graph which schematically explains how the oxygen permeability of the barrier layer 12 varies with the proportion (weight %) of EVOH in the barrier layer. The vertical axis in the graph represents the oxygen permeability ( $O_2$  perm.) while the horizontal axis represents the proportion of EVOH (% EVOH) in the barrier layer. On the right-hand vertical axis there are shown the relative oxygen permeabilities for a number of conventional oxygen barrier materials in the field including Al foil (Al), ethylene/vinyl alcohol copolymer (EVOH), polyethylene terephthalate (PET) and polyvinyl chloride (PVC). Of these known barrier materials Al foil has the lowest oxygen permeability, which in the graph is made equal to 0. The comes EVOH with a corresponding oxygen permeability of c. 0.01, while the two other barrier materials have corresponding oxygen permeability values of c. 6 (PET) and c. 14 (PVC) respectively.

The oxygen permeability for the barrier layer according to the invention varies along a S-shaped curve from a value higher than 14 (with 0% EVOH) to a value of c. 0.09\* (with c. 40% EVOH) which with proportions of EVOH increasing beyond this remains practically constant at this level, i.e. c. 0.09.

In order to give the best possible oxygen-tightness in the material according to the invention, the proportion of EVOH in the barrier layer should thus be c. 50 weight %, at which the barrier layer exhibits the lowest possible oxygen-tight properties\*\* with the use of the minimum possible proportion of EVOH. Acceptable oxygen-tight properties are, however, already achieved with proportions of EVOH of c. 40 weight %, which gives an oxygen barrier better than the oxygen permeability for polyethylene terephthalate (PET). In practice therefore the EVOH content should be c. 40-80 weight %, but preferably c. 50 weight %.

The material 10 according to the invention can be manufactured by extrusion of all layers of material forming part of the material, but is preferably manufactured by co-extrusion with the use of a device of the type which is shown schematically in figure 2. The device with the general reference designation 20 contains 4 extruders coupled together 21, 22, 23 and 24 with a common orifice head 25, and also a cylinder or roller arrangement placed in front of the orifice head, containing cooled cylinders or rollers 26 rotating in the direction of the arrows.

Through the central extruder 21 melted output material for the skeletal layer 11 is propelled forward, i.e. a mixture of the chosen plastic and filler to an amount of between 50 and 80 % of the total weight of the mixture, while the extruders 22 and 23 propel forward melted output material for the barrier layer 12, i.e. a mixture of the same plastic as in the skeletal layer and a plastic of another type, and melted output material for the outer layer 13, i.e. plastic of the type chosen for the skeletal layer, respectively. The extruder 25 propels forward melted output material for the outer layer 14, i.e. plastic of the type chosen for the skeletal layer. The four flows of melted material indicated with dotted lines are pressed through the slot-shaped opening of the orifice head 25 and taken jointly with each other through the pinch between the contra-rotating press cylinders or rollers 26 for cooling and durable joining together of the layers of material through surface fusion of the plastic of the aforesaid same type in the respective neighbouring layers of material to form the finished packaging material 10 in strip form. The packaging material 10 can then be wound on to a supply roller 10'.

In accordance with the present invention it is

\* This appears to be a misprint for 2.16 (Cf. other figures)

\* The graph itself shows that 0.19, printed here<sup>55</sup> is a misprint.

\*\* This seems to be a misprint, with either lowest meant to be best or oxygen-tight properties meant to be oxygen permeability.

thus possible in a simple manner with the use of already existing technology and equipment to produce packaging material in the form of a sheet or strip which exhibits good binding strength between all the layers of material forming part of the material and which has oxygen-tight properties in the same class as a conventional packaging material containing an Al foil as a barrier layer. The packaging material is in addition easy to recover and reuse, since all layers of material contain plastic of the same type, which entails that the material can be shaped into containers with the least possible waste of material, whether the material is shaped through fold forming, thermo-forming or other mechanical processing for shaping.

Even if the invention has been described with particular reference to a single embodiment shown as a preferred example, it should be observed that several modifications obvious for the specialists are possible within the framework of the idea of the invention as defined by the patent claims that follow.

#### Claims

1. Flexible packaging material in the form of a sheet or strip for shaping into form-stable, liquid-tight containers with good oxygen-tight properties through fold forming, thermo-forming or other mechanical processing for shaping of the material, with this packaging material having a stiffening skeletal layer (11) composed of a mixture of plastic and filler, and also a barrier layer (12) attached to one side of the skeletal layer with good adhesion, **characterised by the fact** that the barrier layer (12) is composed of a mixture of plastic of the same type as the plastic in the skeletal layer (11) and a plastic of another type than the one mentioned.
2. Packaging material according to claim 1, **characterised by the fact** that the aforesaid plastic of another type in the barrier layer (12) is an ethylene/vinyl alcohol copolymer.
3. Packaging material according to claim 1 or 2, **characterised by the fact** that the barrier layer (12) is attached to the skeletal layer (11) through surface fusion of the aforesaid plastic of the same type in the barrier layer and skeletal layer respectively.
4. Packaging material according to any of the foregoing claims, **characterised by the fact** that the barrier layer (12) and the skeletal layer (11) are manufactured by extrusion.

5. Packaging material according to any of the claims 1-3, **characterised by the fact** that the barrier layer (12) and the skeletal layer (11) are manufactured by co-extrusion.
6. Packaging material according to any of the claims 2-5, **characterised by the fact** that the amount of ethylene/vinyl alcohol copolymer in the barrier layer (12) is at least 40% of the total weight of the mixture.
7. Packaging material according to any of the foregoing claims, **characterised by the fact** that the barrier layer (12) has an outer layer (13) of plastic of the aforesaid same type attached to the barrier layer.
8. Packaging material according to any of the foregoing claims, **characterised by the fact** that the skeletal layer (11) has an outer layer (14) of plastic of the aforesaid same type.
9. Packaging material according to claim 7 or 8, **characterised by the fact** that the aforesaid outer layer(s) (13, 14) is or are attached to the barrier layer (12) and the skeletal layer (11) through surface fusion of the aforesaid plastic of the same type in the respective neighbouring layers.
10. Packaging material according to any of claims 7-9, **characterised by the fact** that the aforesaid outer layer(s) (13, 14) is or are manufactured through extrusion.
11. Packaging material according to any of claims 7-9, **characterised by the fact** that the aforesaid outer layer(s) (13, 14) is or are manufactured through co-extrusion together with the barrier layer (12) and the skeletal layer (11).
12. Packaging material according to any of the foregoing claims, **characterised by the fact** that the aforesaid plastic of the same type in the barrier layer (12), the skeletal layer (11) and the aforesaid outer layers (13, 14) is composed of a propylene homopolymer or ethylene-/propylene copolymer.
13. Packaging material according to any of the foregoing claims,

characterised by the fact that the filler in the skeletal layer (11) is composed of powdered, granular and/or flaked chalk, talc, mica, clay etc. either each on its own or in any desired combinations with each other.

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14. Packaging material according to any of the foregoing claims,  
characterised by the fact that the filler in the skeletal layer (11) is between 50 and 80% of the total weight of the mixture.

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15. Packaging material according to any of the foregoing claims,  
characterised by the fact that the barrier layer (12) has a thickness of between 5 and 50  $\mu\text{m}$ .

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16. Use of a flexible packaging material in the form of a sheet or strip according to any of the claims 1-15 for manufacture of form-stable, liquid-tight containers with good oxygen-tight properties through fold forming, thermo-forming or other mechanical processing for shaping of the material.

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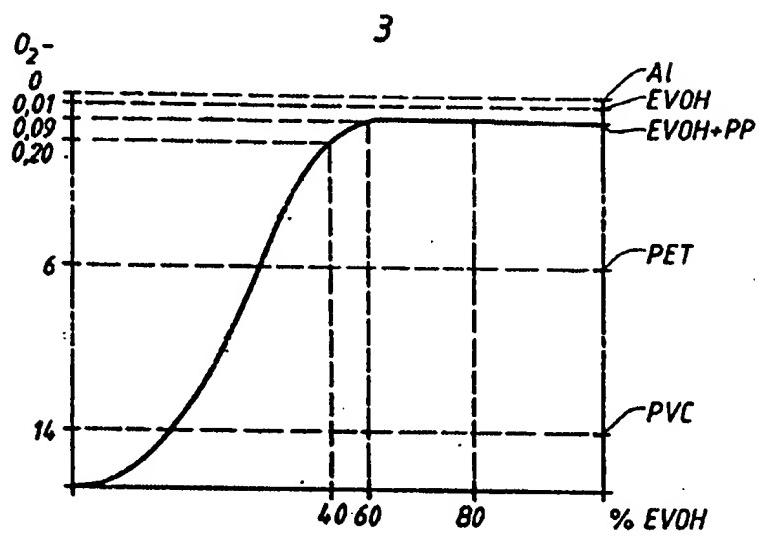
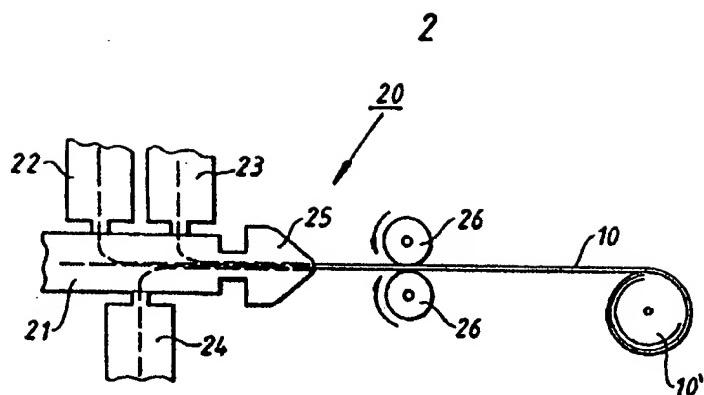
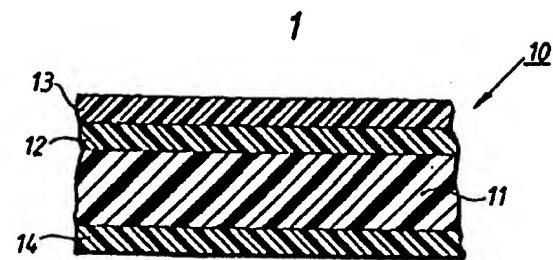
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DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IN C.I.)
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Y	- - -	8-14	
X	GB,A, 1 409 958 (TOYO SEIKAN KAISHA LTD) 15 October 1975 *See page 1, column 2, line 59 - column 2, line 63; page 2, column 2, line 100 - column 2, line 108;*	1-7,15-16	
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Y	GB,A, 1 521 568 (POLYSAR LIMITED) 16 August 1978 *See page 1, column 1, line 36 - column 2, line 65; page 2, column 2, line 88 - column 2, line 98; page 3, column 1, line 54 - column 2, line 59; page 3, column 2, line 107 - column 2, line 112;*	8-14	
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
STOCKHOLM	31-03-1992	Nilsson, J.	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone	T : theory or principle underlying the invention		
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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int'l Cl.)
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			TECHNICAL FIELDS SEARCHED (Int'l Cl.)
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone	T : theory or principle underlying the invention		
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